Heat flow in nanostructures in the Casimir regime

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In small structures the phonon mean free path due to phonon-phonon interactions and defect scattering may exceed the sample dimensions. The thermal conductivity then becomes dependent on the size and shape of the sample. In this article we present calculations of the conductivity under these conditions for several different geometries. Numerical results are presented for Si and GaAs.

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I. INTRODUCTION

In a series of experiments performed between 1935 and 1938 de Haas and Biermasz[1–3] discovered that at low temperatures (typically below about 10 K) the thermal conductivity of high-quality dielectric crystals becomes dependent on the sample size. Peierls,[4] in a letter to de Haas, suggested that this effect might be due to the "reflexion of elastic waves from the walls of the rod." This general idea was worked out in more detail by Casimir in 1938.[5] At low temperatures only long-wavelength acoustic phonons are important. Casimir showed that the conductivity could be written as

$$\kappa = \tfrac{1}{3} C \langle v \rangle \Lambda_C, \tag{1}$$

where C is the specific heat, and $\langle v \rangle$ is an appropriate average phonon velocity. If the sound speed is isotropic and independent of phonon frequency, this is given by

$$\langle v \rangle = \frac{1/v_l^2 + 2/v_t^2}{1/v_l^3 + 2/v_t^3}, \tag{2}$$

where v_l and v_t are the velocities of the longitudinal and transverse phonons, respectively. Casimir found that for a cylindrical rod of radius R the effective mean free path was

$$\Lambda_C = 2R. \tag{3}$$

For a rod of square section with side D the result is

$$\Lambda_C = \frac{D}{2}[3\ln(1+\sqrt{2}) - \sqrt{2} + 1] \approx 1.115D. \tag{4}$$

Note that Eqs. (3) and (4) are based on the assumption that the scattering at the surface of the sample is perfectly diffuse, i.e., a phonon incident on the surface of the sample is reemitted in a random direction.

Equations (3) and (4) hold when the rate of phonon scattering due to phonon-phonon interactions and phonon scattering by defects is so small that phonons can travel across the width of the sample without scattering.[6] For macroscopic samples, this situation occurs only at low temperatures. However, for nanostructures, scattering by boundaries is important at much higher temperatures, and for sufficiently small samples the conductivity will be size dependent even at room temperature. One can make a rough estimate of the size required for this. For example, if for silicon we take the average phonon velocity to be the Debye velocity [5700 m s^{-1} (Ref. 7)], the kinetic formula implies an average mean free path in a bulk sample at room temperature of 47 nm. Thus to be in the Casimir regime the dimensions need to be less than this.

This effect has been studied experimentally[8–15] in a variety of nanostructure samples. Experimental work is complicated by the possible presence of defects within the material and by imperfect knowledge of the details of the sample surface. Most of the theoretical work has used the phonon Boltzmann equation[16–21] with some approximation to the collision term. For example, the scattering of phonons by isotopes has often been taken to be directly proportional to the fourth power of the phonon frequency, although this is in fact correct only for frequencies much below the Debye frequency. In most cases, a simplified form for the phonon dispersion relation has been taken. However, a quantitative calculation of the conductivity needs to take into account the details of the phonon dispersion relation and to allow for phonon focusing effects.[22] Calculations have also been made using molecular dynamics[23] which can include these effects but such calculations have to be corrected for quantum effects.

In this paper, we present calculations of the conductivity under Casimir conditions for several different geometries. We first derive some exact analytical results for the case that the phonon velocity is independent of wave vector. We then present numerical results for silicon and gallium arsenide using a realistic phonon dispersion relation. Throughout this paper we are assuming that the relevant phonon wavelength is much less than the dimensions of the structure, so that the phonon dispersion relation is the same as in bulk.

II. ANALYTICAL RESULTS

We consider heat flow along a rod with axis in the z direction. The rod extends from $z = -\infty$ to $z = \infty$. We take the scattering of the phonons at the walls to be perfectly diffuse so that the walls act as "blackbody" emitters and absorbers of phonons. Thus, the rate at which energy is emitted from a surface element of area dA is given by

$$\frac{1}{8\pi^3} \sum_j \iiint \vec{v}_{\vec{k}j}.\hat{n}\, n_{\vec{k}j}(T) \hbar\omega_{\vec{k}j} k^2\, dk\, d\cos\theta\, d\phi\, dA, \tag{5}$$

where $\hat{n}$ is a unit vector normal inward to the surface, $\vec{v}_{\vec{k}j}$ is the group velocity of a phonon with wave vector $\vec{k}$ and polarization j, and $n_{\vec{k}j}$ is the occupation number for this phonon. The integrals are over all of $\vec{k}$ space subject to the condition that $\vec{v}_{\vec{k}j} \cdot \hat{n}$ be positive. Let $\Lambda_{\vec{k}j}$ be the distance that the phonon $\vec{k}j$ travels in the z direction before being absorbed at the wall of the sample (Fig. 1). In order to derive a general expression for the conductivity, it is convenient to consider a situation in which the temperature distribution along the tube is

$$T(z) = T_0 + \Delta T(z), \tag{6}$$

where $\Delta T(z) = a\,\delta(z)$ and a is some coefficient. Then the net rate of loss of heat occurring from the region close to $z = 0$ is

$$\begin{aligned}\frac{1}{8\pi^3}\oint dl \sum_j \iiint \vec{v}_{\vec{k}j}\cdot\hat{n}\; a\,\left.\frac{\partial n_{\vec{k}j}(T)}{\partial T}\right|_{T_0} \hbar\omega_{\vec{k}j}\,k^2\,dk\,d\cos\theta d\phi \\ = \frac{a\hbar^2}{8\pi^3 k T_0^2}\oint dl \sum_j \iiint \vec{v}_{\vec{k}j}\cdot\hat{n}\; n_{\vec{k}j}(n_{\vec{k}j}+1)\,\omega_{\vec{k}j}^2\,k^2\,dk\,d\cos\theta\,d\phi,\end{aligned} \tag{7}$$

where the line integral is around the circumference of the rod, and we used the relation

$$\left.\frac{\partial n_{\vec{k}j}}{\partial T}\right|_{T_0} = \frac{\hbar\,\omega_{\vec{k}j}}{kT_0^2} n_{\vec{k}j}(n_{\vec{k}j}+1). \tag{8}$$

A phonon leaving the position $z = 0$ with wave vector $\vec{k}$ and polarization j will be absorbed at the position $z = \Lambda_{\vec{k}j}$ and will contribute to the increase in the heat at this position. Thus, if $Q(z)$ is the heat per unit distance at position z, we have

$$\begin{aligned}\frac{dQ(z)}{dt} = \frac{a\hbar^2}{8\pi^3 kT_0^2}\oint dl \sum_j \iiint \vec{v}_{\vec{k}}\cdot\hat{n}\, n_{\vec{k}j}(n_{\vec{k}j}+1) \\ \times\,\omega_{\vec{k}j}^2\,\delta(z-\Lambda_{\vec{k}j})\,k^2\,dk\,d\cos\theta\,d\phi.\end{aligned} \tag{9}$$

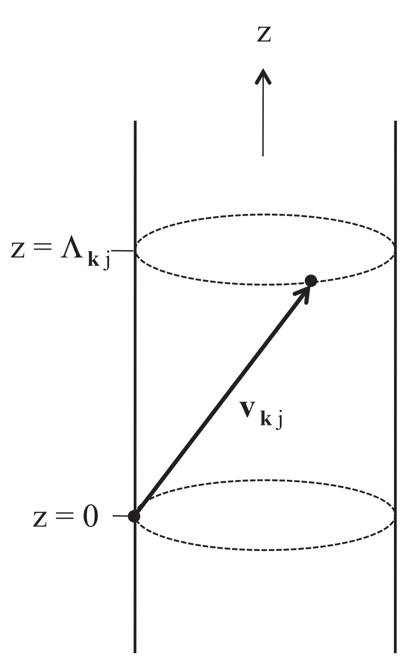

FIG. 1. Schematic of a rod in which a phonon leaves a point on the wall at $z = 0$ and travels with group velocity $\vec{v}_{\vec{k}j}$ until it hits another point on the wall with $z = \Lambda_{\vec{k}j}$.

From this it follows that

$$\begin{aligned}\frac{d}{dt}\int_{-\infty}^{\infty} Q(z)z^2dz = \frac{a\hbar^2}{8\pi^3kT_0^2}\oint dl \sum_j \iiint \vec{v}_{\vec{k}j}\cdot\hat{n}\,n_{\vec{k}j} \\ \times (n_{\vec{k}j}+1)\Lambda_{\vec{k}j}^2\,\omega_{\vec{k}j}^2\,k^2dk\,d\cos\theta\,d\phi.\end{aligned} \tag{10}$$

We now derive an expression for the same derivative based on the macroscopic equations governing heat transport. Consider a plane which lies perpendicular to the direction of the rod and passing through the position z. Let $J(z)$ be the rate at which heat flows across this plane per unit time. From Fourier's law,

$$J(z) = -\kappa A\frac{\partial \delta T}{\partial z}, \tag{11}$$

where $\delta T(z) = T(z) - T_0$, and A is the cross-sectional area of the rod. But

$$\frac{\partial\, Q(z)}{\partial\, t} = -\frac{\partial J}{\partial z}. \tag{12}$$

Therefore

$$\frac{\partial Q(z)}{\partial t} = \kappa A\frac{\partial^2\delta T}{\partial z^2}. \tag{13}$$

Hence,

$$\begin{aligned}\frac{d}{dt}\int_{-\infty}^{\infty} Q(z)z^2dz &= \kappa A\int_{-\infty}^{\infty}\frac{\partial^2\delta T(z)}{\partial z^2}z^2dz \\ &= -2\kappa A\int_{-\infty}^{\infty}\frac{\partial\delta T(z)}{\partial z}zdz \\ &= 2\kappa A\int_{-\infty}^{\infty}\delta T(z)\,dz.\end{aligned} \tag{14}$$

In this derivation the fact that the δT is zero at $z = \pm\infty$ is used. From conservation of energy, the last integral on the right-hand side of this equation must be a constant. It follows that we can use the initial condition [Eq. (6)] to evaluate this integral. Therefore

$$\frac{d}{dt}\int_{-\infty}^{\infty} Q(z)\, z^2\, dz = 2\, A\, \kappa\, a. \tag{15}$$

Then by comparing Eqs. (10) and (15) we obtain

$$\kappa = \frac{\hbar^2}{16\pi^3 k T_0^2 A}\oint dl \sum_j \iiint \vec{v}_{\vec{k}j}\cdot\hat{n}\, n_{\vec{k}j}(n_{\vec{k}j}+1) \times \Lambda_{\vec{k}j}^2\, \omega_{\vec{k}j}^2 k^2\, dk\, d\cos\theta\, d\phi. \tag{16}$$

We first consider the results from this formula if the phonons all have the same speed v, regardless of polarization, propagation direction, or magnitude of the wave vector. The generalization to allow for different velocities of longitudinal and transverse phonons, but still retaining isotropy, is straightforward. The Casimir mean free path is given as

$$\Lambda_C = 3\kappa/Cv. \tag{17}$$

For cylindrical and square cross-section rods, Eqs. (16) and (17) lead to the results obtained by Casimir [Eqs. (3) and (4)].

It is straightforward but tedious to find κ for a rectangular plate with sides D (thickness) and W (width) and infinite length along the z direction. The result is

$$\Lambda_C = \frac{Dn^{1/2}}{4}\left\{\begin{array}{l} 3n^{1/2}\ln[n^{-1}+(n^{-2}+1)^{1/2}] + 3n^{-1/2}\ln[n+(n^2+1)^{1/2}] \\ -(n+n^3)^{1/2} + n^{3/2} - (n^{-1}+n^{-3})^{1/2} + n^{-3/2} \end{array}\right\}, \tag{18}$$

where $n = W/D$. This result has been obtained previously by McCurdy *et al.*[7] The Casimir mean free path has a logarithmic divergence when the width of the plate goes to infinity. The limiting form is

$$\Lambda_C = \tfrac{3}{4}D\left[\ln 2 + \tfrac{1}{2} + \ln(W/D)\right] = D\,[0.895 + 0.75\ln(W/D)]\,. \tag{19}$$

This divergence comes from phonons which propagate nearly in the plane of the plate, i.e., in the plane normal to the thickness, and which can therefore have very large values of $|\Lambda_{\vec{k}j}|$. The variation of Λ_C with the width of the plate is shown in Fig. 2. The result Eq. (18) shows that when the Casimir condition holds, the thermal conductivity for heat flow along a plate cannot be considered to be a function of just the thickness of the plate; the width also plays a role. It has been claimed that the Casimir length for heat flow along a plate is equal to twice the thickness.[19] This appears to be incorrect.

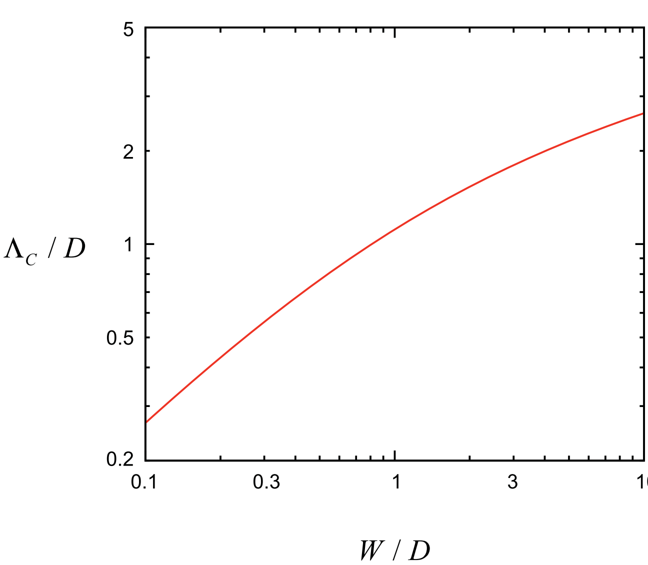

FIG. 2. (Color online) Calculated Casimir length for heat flow along a rectangular plate with cross-sectional dimensions D and W. Results are for an isotropic solid in which all phonons have the same velocity.

It is interesting to consider what happens to this logarithmic divergence when the plate is curved. There are two cases to consider. First, consider a plate which has infinite width and which is bent along the direction in which heat is flowing. Thus, the direction of heat flow is slowly changing with position. One can formulate this by a method similar to the approach used to derive Eq. (16). We suppose that the plate is bent so that the outer radius is r_1 and the inner radius is r_2. We can consider that the plate is part of an annulus. Then it can be shown that the conductivity is given by

$$\kappa = \frac{\hbar^2}{16\pi^3 k T_0^2 \ln(r_1/r_2)} \times \left\{\begin{array}{l} r_1\sum_j \iiint \vec{v}_{\vec{k}j}\cdot\hat{n}\ \omega_{\vec{k}j}^2\, n_{\vec{k}j}(n_{\vec{k}j}+1)\,\theta_{\vec{k}j}^2\, d^3\vec{k} \\ +r_2\sum_j \iiint \vec{v}_{\vec{k}j}\cdot\hat{n}\ \omega_{\vec{k}j}^2\, n_{\vec{k}j}(n_{\vec{k}j}+1)\theta_{\vec{k}j}^2\, d^3\vec{k} \end{array}\right\}, \tag{20}$$

where $\theta_{\vec{k}j}$ is the angle by which the phonon $\vec{k}j$ moves around the annulus, and the remaining notation is the same as in Eq. (16). We have not been able to perform analytically the integrals that enter in this expression for the conductivity, but for large values of the radius of curvature R the numerical result is well approximated by the expression

$$\Lambda_C = D\,[1.29 + 0.385\ln(R/D)]\,. \tag{21}$$

As $R \to \infty$, Λ_C diverges logarithmically as expected from the formula for the flat plate [Eq. (19)].

As a second geometry, we consider heat flow in a plate which is curved around the direction of heat flow, i.e., the z axis. This geometry can be considered to be a special case of a more general problem, namely the flow of heat along a hollow cylinder.[24] Let the outer and inner radii of the cylinder be r_1 and r_2, respectively. Consider a phonon leaving the outer

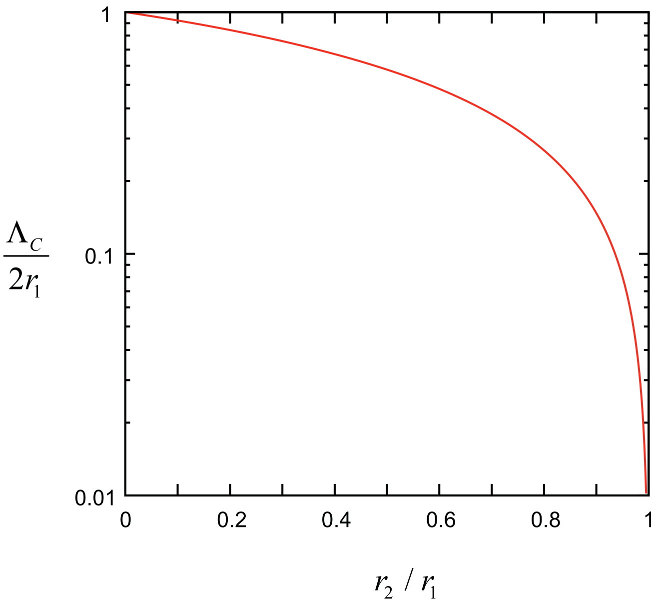

FIG. 3. (Color online) Calculated Casimir length for heat flow along a cylinder of radius r_1 with a hole of diameter r_2. Results are for an isotropic solid in which all phonons have the same group velocity.

surface at a position $(r_1, 0, 0)$ with direction θ,ϕ. This phonon will strike the inner wall if $r_2 > r_1|\sin\phi|$, in which case

$$\Lambda_{\vec{k}j} = \left[r_1\cos\phi - \left(r_2^2 - r_1^2\sin^2\phi\right)^{1/2}\right]\cot\theta. \tag{22}$$

If $r_2 < r_1|\sin\phi|$ the phonon hits the outer wall and

$$\Lambda_{\vec{k}j} = 2r_1\cos\phi\cot\theta. \tag{23}$$

A phonon leaving the inner wall always hits the outer wall and

$$\Lambda_{\vec{k}j} = \left[\left(r_1^2 - r_2^2\sin^2\phi\right)^{1/2} - r_2\cos\phi\right]\cot\theta. \tag{24}$$

Substituting these results into Eq. (16) and performing the integrals over θ and ϕ, we obtain

$$\Lambda_C = \frac{2r_1}{1-\beta^2}\left\{1 - \frac{3\beta}{4} + \frac{3\beta^3}{4} - \frac{1}{2}[(1+\beta^2)E(\beta^2) - (1-\beta^2)K(\beta^2)]\right\}, \tag{25}$$

where $\beta = r_2/r_1$, and K and E are complete elliptic integrals of the first and second kind. Figure 3 shows Λ_C as a function of r_2/r_1. We can use this general expression to find the conductivity of a thin curved plate of thickness D and radius of curvature R. To do this we set $r_1 = R + D/2$, and $r_2 = R - D/2$ and take the limit of Eq. (25) when $D \ll R$. After some algebra the result is that

$$\Lambda_C = D\left(\tfrac{21}{16} + \tfrac{3}{8}\ln 8 + \tfrac{3}{8}\ln R/D\right) = D(2.092 + 0.375\ \ln R/D). \tag{26}$$

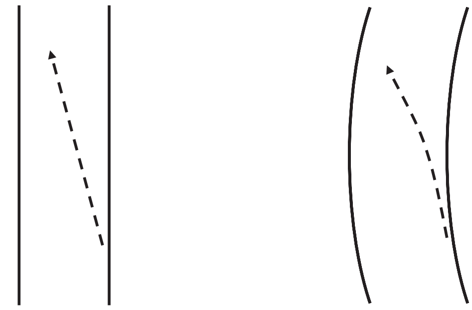

FIG. 4. Schematic diagram showing the effect of bending a plate on phonon propagation. The solid line represents the walls of the plate and the dashed line shows a possible trajectory of a phonon.

The derivation uses the series expansions[25]

$$E(\beta^2) = 1 + \left(\frac{1}{2}\ln\frac{4}{\gamma} - \frac{1}{4}\right)\gamma^2 + \left(\frac{3}{16}\ln\frac{4}{\gamma} - \frac{13}{64}\right)\gamma^4\ldots,$$
$$K(\beta^2) = \ln\frac{4}{\gamma} + \left(\frac{1}{4}\ln\frac{4}{\gamma} - \frac{1}{4}\right)\gamma^2 + \left(\frac{9}{64}\ln\frac{4}{\gamma} - \frac{21}{128}\right)\gamma^4\ldots, \tag{27}$$

which hold when $\gamma \equiv \sqrt{1-\beta^2}$ is very small.

From Eqs. (19), (21), and (26) it follows that it may be possible to make changes to the thermal conductivity of a plate by bending. However, it is not simple to make a reliable estimate of the change that can be made. The difficulty is that the results we have obtained hold strictly in the limit that there is no scattering other than at the boundaries. It is only if this is true that the conductivity becomes infinite when there is no curvature. Phonon scattering within the volume of the material will cut off the logarithmic divergence. A second consideration is that when a plate is bent, an inhomogeneous static strain will be set up. There will be a compressive strain on the concave side of the plate and an expansion on the other. This will have the consequence that the phonon dispersion relation will now depend on position, and a phonon crossing the plate will have a propagation direction which changes continuously as it moves (mirage effect Fig. 4). Since sound velocity usually increases under compression, this effect enhances the change in the conductivity that occurs just due to a change in the geometry.

III. NUMERICAL EXAMPLES

We have performed a numerical evaluation of the conductivity for Si and GaAs. The calculations use the models of the phonon dispersion relation described in Refs. 26 and 27. These models reproduce the measured dispersion relations to within a few percent. To perform the integral in Eq. (16), we have used a large number of randomly selected wave vectors, typically 300 000. In Figs. 5 and 6 we show results for the thermal conductivity of plates of Si and GaAs. The plate thickness is chosen to be 30 nm. For plates with the same ratio of thickness D to width W, the conductivity must be proportional to D. It follows that the conductivity must satisfy the relation

$$\kappa(D,W) = \frac{W}{D}\kappa(D, D^2/W), \tag{28}$$

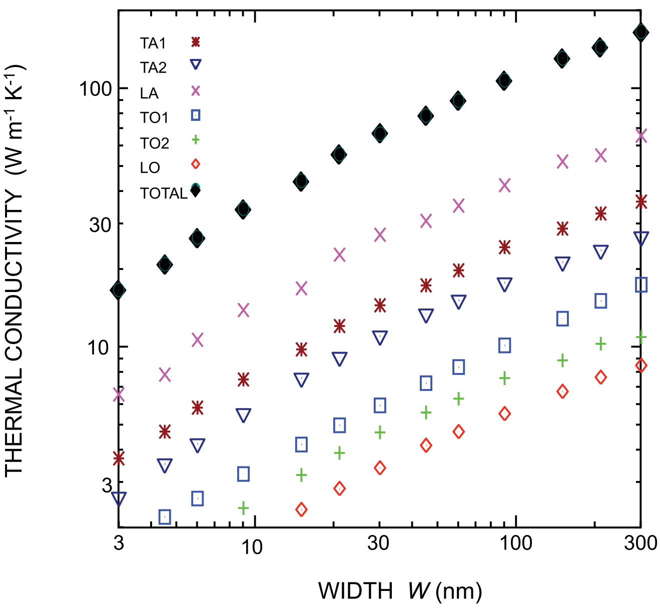

FIG. 5. (Color online) Calculated thermal conductivity κ for heat flow along a rectangular plate as a function of the width W. The thickness D of the plate is 30 nm and the results are for silicon at 300 K. The contributions from transverse acoustic, longitudinal acoustic, and optical phonons are shown separately.

and this provides a simple check on the accuracy of the calculations.

The largest contribution to the thermal conductivity comes from the longitudinal acoustic phonons. As can be expected, the optical phonons make a very small contribution. For a square cross-section rod of Si the conductivity is $66\ \mathrm{W\,m^{-1}\,K^{-1}}$. The specific heat of Si is $1.66 \times 10^6\ \mathrm{J\,m^{-3}\,K^{-1}}$. Thus, in order for Eq. (1) together with Eq. (4) to describe

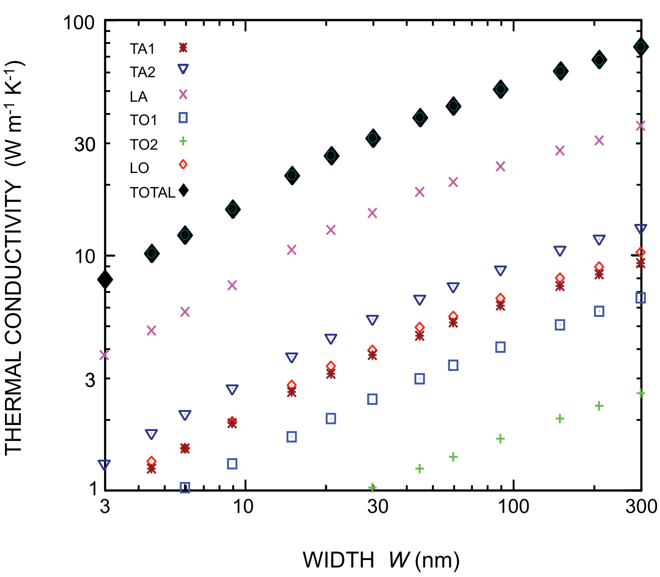

FIG. 6. (Color online) Calculated thermal conductivity κ for heat flow along a rectangular plate as a function of the width W. The thickness D of the plate is 30 nm and the results are for gallium arsenide at 300 K. The contributions from transverse acoustic, longitudinal acoustic, and optical phonons are shown separately.

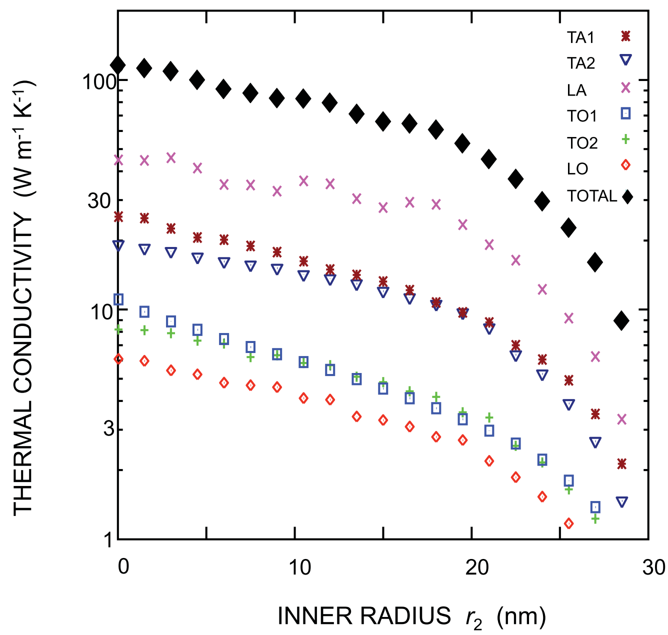

FIG. 7. (Color online) Calculated thermal conductivity κ for heat flow along a cylinder of radius $r_1 = 30$ nm with a hole of radius r_2. Results are for silicon at 300 K with the cylinder axis along the [100] direction. The contributions from transverse acoustic, longitudinal acoustic, and optical phonons are shown separately.

the results of the numerical calculations, the effective average velocity entering in Eq. (1) has to be $\langle v \rangle = 3600\ \mathrm{m\,s^{-1}}$. As a result of phonon dispersion, this velocity is considerably less than the sound velocity in Si. For example, the longitudinal and transverse sound velocities in the [100] direction of Si are

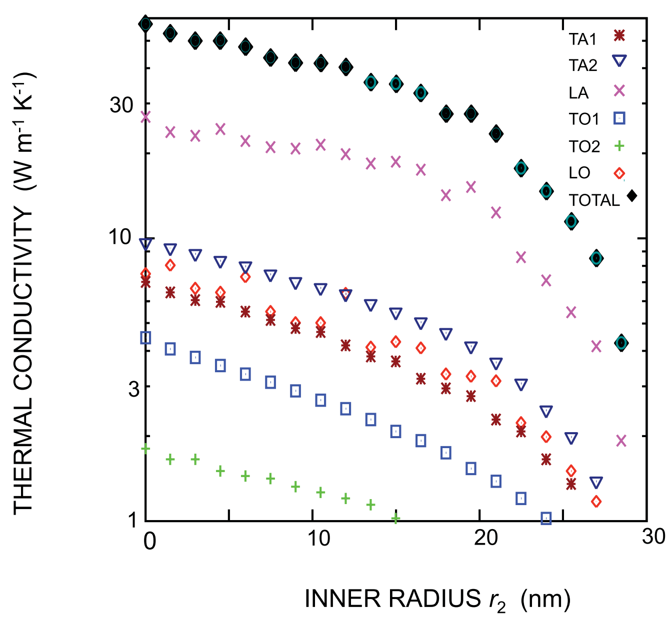

FIG. 8. (Color online) Calculated thermal conductivity κ for heat flow along a cylinder of radius $r_1 = 30$ nm with a hole of radius r_2. Results are for gallium arsenide at 300 K with the cylinder axis along the [100] direction. The contributions from transverse acoustic, longitudinal acoustic, and optical phonons are shown separately.

8400 m s^{-1} and 5200 m s^{-1}, respectively. The corresponding average velocity for GaAs is found to be 1700 m s^{-1}.

We emphasize that the results shown in Figs. 5 and 6 are based on the assumption that there is negligible phonon scattering in the bulk of the material. The bulk thermal conductivity of Si is 141 W m^{-1} K^{-1}, i.e., about twice the value that we find for a square rod of side 30 nm. This therefore indicates that to a reasonable approximation, structures with lateral dimensions of less than or equal to 30 nm are in the Casimir regime. For GaAs the bulk conductivity is 55 W m^{-1} K^{-1} and for the same geometry the calculated value is 35 W m^{-1} K^{-1}, and so again a 30 nm structure should be in the Casimir regime.

Results for the thermal conductivity for heat flow along the axis of a hollow cylinder with outer radius $r_1 = 30$ nm are shown in Figs. 7 and 8. The largest contribution again comes from the longitudinal acoustic phonons. The conductivity of the solid cylinder is found to be 115 W m^{-1} K^{-1} for Si and 57 W m^{-1} K^{-1} for GaAs. These values are close to the values of the bulk conductivity for these materials; this suggests that cylinders with radius less than 30 nm are in the Casimir regime.

It is interesting to compare the results obtained here with a calculation by Mingo for silicon nanowires.[28] He used a realistic interatomic potential to calculate the phonon modes in the wire, i.e., he did not assume that the phonon dispersion was the same as in the bulk. In addition, he included contributions to the scattering rate from anharmonicity and isotope scattering. The phonon scattering time τ was assumed to depend only on frequency. For the smallest radius nanowire (18.5 nm) a conductivity at room temperature of 17 W m^{-1} K^{-1} was obtained. This compares with the result of 70 W m^{-1} K^{-1} that follows from our calculation for the same geometry. It is possible that the difference arises because a wire of this diameter is not fully in the Casimir regime; our value of 70 W m^{-1} K^{-1} is based on the approximation that it is. It would be interesting to investigate this further.

IV. SUMMARY

In summary, we have presented quantitative calculations of the phonon thermal conductivity in the boundary scattering (Casimir) regime. Our numerical results for Si and GaAs are based on accurate phonon dispersion relations. The results obtained assume that the scattering at the sample surface is diffuse.

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